

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: Michel et al.

Docket: 1999DE132

Serial No.: 09/722,760

Group Art Unit: 1753

Filed: 11/27/2000

Examiner: Dote, Janis L.

For: Use of Salt-Like Structured Silicas as Control Agents

SUPPLEMENTAL AMENDMENTAssistant Commissioner of Patents
Washington, D.C. 20231

Dear Sir:

In Response to a telephone discussion with Examiner Dote on April 25, 2003,
please amend the Application as follows:

IN THE CLAIMS

1. (Previously Amended) A method of imparting, controlling or improving the charge of an electrophotographic toner or developer, or an electret material, comprising the step of adding a structured silicate salt in which the cation is a low molecular weight organic cation and the anion is an island, cyclic, group, chain, ribbon, laminar or matrix silicate or a combination thereof to a binder of an electrophotographic toner or developer or of an electret material.

2) (Cancelled)

3) (Cancelled)

CERTIFICATE OF MAILING/TRANSMISSION (37 CFR 1.8a) and 1.10

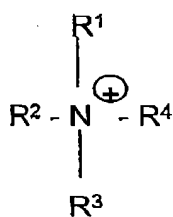
I hereby certify that this correspondence is, on the date shown below,
being transmitted by facsimile to the Assistant Commissioner for Patents,
Attention Examiner Dote, Janis J. to Fax No. (703) 305-6078 (10 pages)

Vicki L. Sgro: Date: April 25, 2003

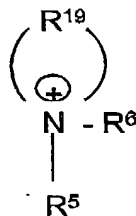
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4) (Original) The method as claimed in claim 1, wherein the low molecular weight organic cation is a substituted ammonium, phosphonium, thionium or triphenylcarbonium ion or a cationic metal complex.

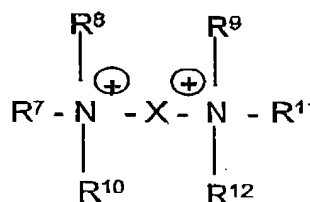
5) (Previously Amended) The method as claimed in claim 4, wherein the ammonium ion has one of the formulae (a) - (j)



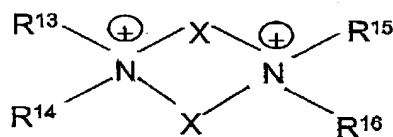
(a)



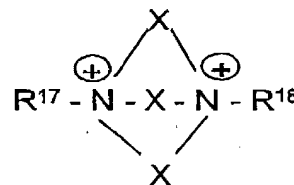
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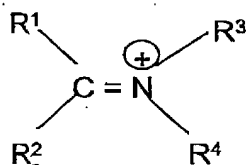
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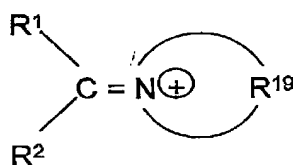
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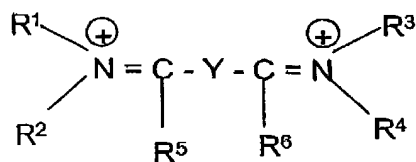


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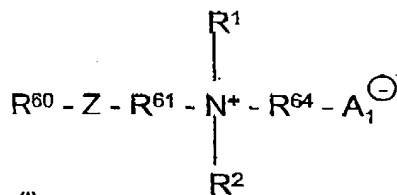


(g)

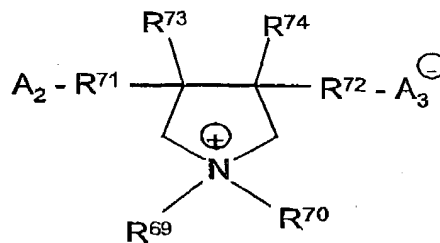
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(h)



(i)

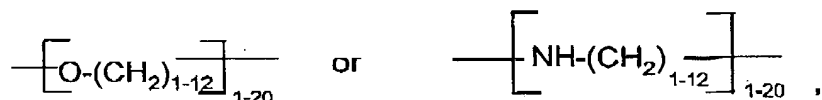


(j)

in which

R¹ to R¹⁸ are identical or different and represent hydrogen, CN, (CH₂)₁₋₁₈CN, halogen, branched or unbranched C₁-C₃₂-alkyl, mono- or polyunsaturated C₂-C₃₂-alkenyl, C₁-C₂₂-alkoxy, C₁-C₂₂-hydroxyalkyl, C₁-C₂₂-halogenoalkyl, C₂-C₂₂-halogenoalkenyl, C₁-C₂₂-aminoalkyl, (C₁-C₁₂)-trialkyl-ammonium-(C₁-C₂₂)-alkyl; (C₁-C₂₂)-alkylene-(C=O)O-(C₁-C₃₂)-alkyl, (C₁-C₂₂)-alkylene-(C=O)O-aryl, (C₁-C₂₂)-alkylene-(C=O)NH-(C₁-C₃₂)-alkyl, (C₁-C₂₂)-alkylene-(C=O)NH-aryl, (C₁-C₂₂)-alkylene-O(CO)-(C₁-C₃₂)-alkyl, (C₁-C₂₂)-alkylene-O(CO)-aryl, (C₁-C₂₂)-alkylene-NH(C=O)-(C₁-C₃₂)-alkyl, (C₁-C₂₂)-alkylene-NHCO-aryl,

wherein



are optionally inserted into the acid ester or acid amide bonds;

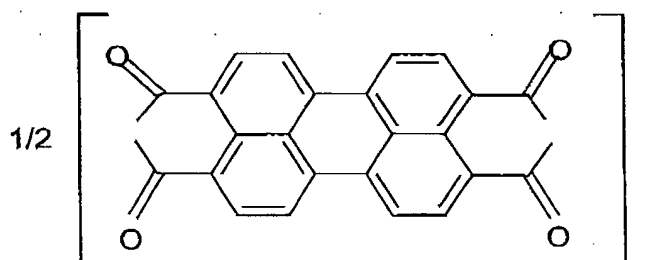
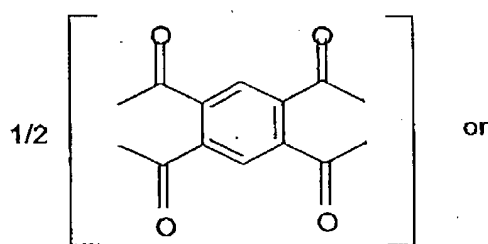
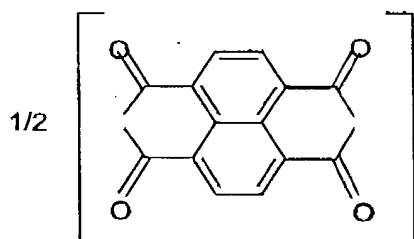
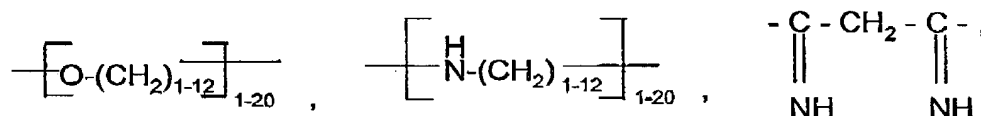
[(C₁-C₁₂)-alkylene-O-]₁₋₁₀₀-H; aryl, (C₁-C₁₈)-alkylenearyl; -(O-SiR'₂)₁₋₃₂-O-SiR'₃, in which R' has the meaning C₁-C₁₂-alkyl, phenyl, benzyl or C₁-C₁₂-alkoxy; heterocyclyl, C₁-C₁₈-alkylene-heterocyclyl, wherein the aryl and heterocyclyl radicals are optionally mono- or polysubstituted on carbon atoms or heteroatoms by C₁-C₁₂-alkyl, C₁-C₄-alkenyl, C₁-C₄-alkoxy, hydroxy-(C₁-C₄)-alkyl, amino-(C₁-C₄)-alkyl, C₁-C₄-alkylimino,

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carboxyl, hydroxyl, amino, nitro, cyano, halogen, C₁-C₁₂-acyl, C₁-C₄-halogenoalkyl, C₁-C₄-alkylcarbonyl, C₁-C₄-alkylcarbonyloxy, C₁-C₄-alkoxycarbonyl, C₁-C₄-alkylaminocarbonyl, C₁-C₄-alkylcarbonylimino, C₆-C₁₀-arylcarbonyl, aminocarbonyl, aminosulfonyl, C₁-C₄-alkylaminosulfonyl, phenyl, naphthyl, or heteroaryl[.];

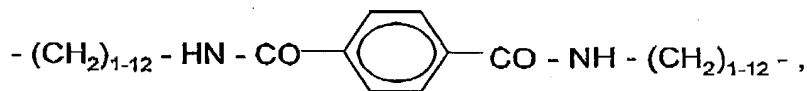
R¹⁹ represents C₄-C₁₁-alkylene, -(C₂H₄-O)₁₋₁₇-(CH₂)₁₋₂-, -(C₂H₄-NR-)₁₋₁₇-(CH₂)₁₋₂-, in which R is hydrogen or C₁-C₁₂-alkyl;

X has the meaning of Y or -CO-CH₂-CO-,



Y has the meaning $\begin{array}{c} -\text{C}- \\ \parallel \\ \text{O} \end{array}$, $\begin{array}{c} -\text{C}- \\ \parallel \\ \text{S} \end{array}$, $\begin{array}{c} -\text{C}- \\ \parallel \\ \text{NH} \end{array}$, -(CH₂)₁₋₁₈-,

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or o-, p-, m-(C₆-C₁₄)-arylene or (C₄-C₁₄)-heteroarylene with 1, 2, 3 or 4 heteroatoms selected from the group consisting of N, O, S and a combination thereof;

R⁶⁰ represents C₁-C₃₂-acyl, C₁-C₂₂-alkyl, C₂-C₂₂-alkenyl, C₁-C₁₈-alkylene-C₆-C₁₀-aryl, C₁-C₂₂-alkylene-heterocyclyl, C₆-C₁₀-aryl or (C₄-C₁₄)-heteroaryl with 1, 2, 3 or 4 heteroatoms selected from the group consisting of N, O, S, and a combination thereof;

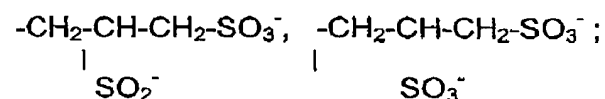
R⁶¹ and R⁶⁴ represent -(CH₂)₁₋₁₈-, C₁-C₁₂-alkylene-C₆-C₁₀-arylene, C₆-C₁₀-arylene, C₀-C₁₂-alkylene-heterocyclyl;

Z represents -NH- or -O-;

A₁⁻ and A₃⁻ represent -COO⁻, -SO₃⁻, -OSO₃⁻, -SO₂⁻, -COS⁻ or -CS₂⁻;

A₂ represents -SO₂Na, -SO₃Na, -SO₂H, -SO₃H or hydrogen;

R⁶⁹ and R⁷⁰ independently of one another represent hydrogen, C₁-C₃₂-alkyl, in which the alkyl chain optionally contain one or more of the groups -NH-CO-, -CO-NH-, -CO-O- or -O-CO-; C₁-C₁₈-alkylene-aryl, C₀-C₁₈-alkylene-heterocyclyl, C₁-C₁₈-hydroxyalkyl, C₁-C₁₈-halogenoalkyl, aryl, -(CH₂)₃-SO₃⁻,



R⁷¹ and R⁷² represent -(CH₂)₁₋₁₂-; and

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R^{73} and R^{74} represent hydrogen or C_1 - C_{22} -alkyl.

6) (Original) The method as claimed in claim 5, wherein R^1 to R^{18} denote hydrogen, CN, CH_2 -CN, CF_3 , C_1 - C_{22} -alkyl, C_2 - C_{18} -alkenyl, C_1 - C_{18} -alkoxy, C_1 - C_{18} -hydroxy-alkyl, C_1 - C_{18} -halogenoalkyl, C_2 - C_{18} -halogenoalkenyl, C_1 - C_{18} -aminoalkyl, (C_1-C_6) -trialkylammonium- (C_1-C_{18}) -alkyl, (C_1-C_{18}) -alkylene- $O(C=O)-(C_1-C_{22})$ alkyl, (C_1-C_{18}) -alkylene- $O(C=O)$ -phenyl, (C_1-C_{18}) -alkylene-NHCO- (C_1-C_{22}) alkyl, (C_1-C_{18}) -alkylene-NHCO-phenyl, (C_1-C_{18}) -alkylene- $(C=O)O-(C_1-C_{22})$ alkyl, (C_1-C_{18}) -alkylene- $(C=O)O$ -phenyl, (C_1-C_{18}) alkylene- $(C=O)NH-(C_1-C_{22})$ alkyl, (C_1-C_{18}) -alkylene-CONH-phenyl, benzyl, phenyl, naphthyl, C_1 - C_{12} -alkylene-heterocyclyl;

R^{19} denotes C_4 - C_5 -alkylene, $-(C_2H_4-O)_{1-9}-(CH_2)_{1-2}$ - or $-(C_2H_4-NH)_{1-9}-(CH_2)_{1-2}$ -;

R^{60} denotes C_1 - C_{18} -acyl, C_1 - C_{18} -alkyl, C_2 - C_{18} -alkenyl, C_1 - C_{12} -alkylene-phenyl, C_1 - C_{18} -alkylene-pyridyl, phenyl or pyridyl;

R^{61} and R^{64} denote $-(CH_2)_{1-12}$ -, C_1 - C_8 -alkylene-phenylene, phenylene or C_1 - C_8 -alkylenepyridylene or piperidylene;

R^{71} and R^{72} denote $-(CH_2)_{1-8}$ and

R^{73} and R^{74} denote hydrogen or (C_1-C_{18}) -alkyl.

7) (Previously Amended) The method as claimed in claim 4, wherein the ammonium ion is an aliphatic or aromatic 5- to 12-membered heterocyclic radical with 1 to 4 atoms selected from the group consisting of N, O and S, or a combination thereof, belonging to the rings.

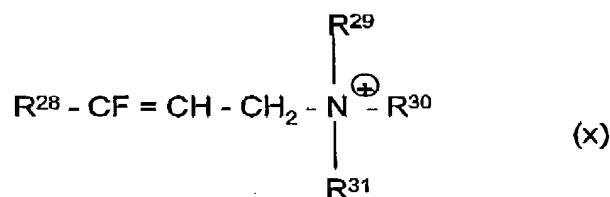
8) (Original) The method as claimed in claim 7, wherein the heterocyclic radical is pyridinium, pyridazinium, pyrimidinium, pyrazinium, purinium, tetraazaporphyrinium, piperidinium, morpholinium, tetrazonium, triaza-cyclononanium or tetraaza-cyclododecanium.

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9) (Original) The method as claimed in claim 4, wherein the cationic metal complex is a metal carboxylate, metal salicylate, metal sulfonate, 1:1 metal-azo complex or a metal dithiocarbamate.

10) (Previously Amended) The method as claimed in claim 9, wherein the metal is selected from the group consisting of Al, Mg, Ca, Sr, Ba, TiO, VO, Cr, V, Ti, Zr, Sc, Mn, Fe, Co, Ni, Cu, Zn and ZrO.

11) (Original) The method as claimed in claim 1, wherein the organic cation is a fluorinated ammonium ion of the formula (x)



in which

R^{28} denotes perfluorinated alkyl having 5 to 11 carbon atoms and

R^{29} , R^{30} and R^{31} are identical or different and denote alkyl having 1 to 5 carbon atoms.

12) (Original) Salt-like structured silicate, in which the silicate is hectorite, beidellite, illite, muscovite, xantophyllite, margarite, sepiolite, saponite, mica, feldspar, nontronite, montmorillonite, smectite, bentonite, faujasite, zeolite A, X or Y, permutite, sasil or a combination thereof; and the cation is an ion of the formula (x) as claimed in claim 9.

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13) (Original) A process for the preparation of a salt-like structured silicate as claimed in claim 12, which comprises combining the silicate and a salt of the cation of formula (x) in an aqueous medium.

14) (Previously Amended) An electrophotographic toner comprising 30 to 99.99% by weight of a binder, and 0.01 to 50% by weight, of at least one salt of ionic structured silicates in which the cation is a low molecular weight organic cation and the anion is an island, cyclic, group, chain, ribbon, laminar or matrix silicate or a combination thereof, based on the total weight of the electrophotographic toner.

15) (Previously Amended) An electrophotographic toner as claimed in claim 14, comprising 40 to 99.5% by weight of a binder, and 0.05 to 20% by weight of at least one salt of ionic structured silicates in which the cation is a low molecular weight organic cation and the anion is an island, cyclic, group, chain, ribbon, laminar or matrix silicate or a combination thereof, based on the total weight of the electrophotographic toner.

16. (Previously Added) The method of claim 4, wherein the ammonium ion is an aliphatic or aromatic 5- to 12-membered heterocyclic radical with 1 to 4 atoms selected from the group consisting of N, O and S, or a combination thereof, belonging to the rings, wherein 2 to 8 rings are fused.

17. (Previously Added) The method as claimed in claim 9, wherein the metal is selected from the group consisting of Al, Mg, Ca, Sr, Ba, TiO, VO, Cr, V, Ti, Zr, Sc, Mn, Fe, Co, Ni, Cu, Zn and ZrO, and the metal complex contains one or more further ligands.

18. (Currently Amended) A method of imparting, controlling or improving the charge of an electrophotographic toner or developer, of a powder coating, or of an electret material, comprising the steps of adding a salt structured silicate in which the cation

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is NH_4^+ , H_3O^+ , an alkali metal, alkaline earth metal, earth metal or transition metal ion or a low molecular weight organic cation or a combination thereof and the anion is an island, cyclic, group, chain, ribbon, laminar or matrix silicate or a combination thereof to a binder of an electrophotographic toner or developer or of a powder coating, or to an electret material.

19. (Previously Amended) An electrophotographic toner or developer comprising distearyldimethyl ammonium bentonite.

20. (Previously Added) The electrophotographic toner as claimed in claim 14, further comprising 0.001 to 50% by weight, of a coloring agent, based on the total weight of the electrophotographic toner.

21. (Previously Amended) A composition comprising 30 to 99.99% by weight of a binder, and 0.01 to 50% by weight, of at least one salt of ionic structured silicates in which the cation is a low molecular weight organic cation and the anion is an island, cyclic, group, chain, ribbon, laminar or matrix silicate or a combination thereof, based on the total weight of the composition, wherein the composition is an electrophotographic toner.

22. (Previously Added) A method of imparting, controlling or improving the charge of an electrophotographic toner or developer, or an electret material comprising the step of adding a distearyldimethyl ammonium bentonite to a binder of an electrophotographic toner or developer or of a powder coating or of an electret material.